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<p><b>(54) Title: ADHESION PROMOTER FOR LINEAR POLYOLEFIN</b></p> <p><b>(57) Abstract</b></p> <p>The present invention is a laminate comprising: a) a first substrate comprising a first substantially linear olefin copolymer; b) an adhesion promoter superposing the first substrate, which adhesion promoter comprises a grafted copolymer of a second substantially linear olefin copolymer; c) a tackifying resin admixed with the adhesion promoter, or an adhesive superposing the adhesion promoter or admixed therewith; and d) a second substrate adhesively bonded to the first substrate. The present invention is also an adhesion promoter comprising a mixture of a copolymer derived from a C<sub>2</sub>-C<sub>20</sub> olefin and an ethylenically unsaturated carboxylic acid or acid anhydride, and a polar group functionalized substantially linear olefin copolymer or a polar group functionalized copolymer of ethylene and a C<sub>3</sub>-C<sub>20</sub> <math>\alpha</math>-olefin having a narrow molecular weight distribution, a random distribution of comonomer units along the polymer backbone, and a homogeneity index of at least 75. The laminate and adhesion promoter of the present invention are useful for a variety of end-use applications, including bathroom scales, shoe soles, carpet-backing, and automobile windshields.</p>			

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## ADHESION PROMOTER FOR LINEAR POLYOLEFIN

This invention relates to an adhesion promoter for a laminate comprising a substantially linear polyolefin elastomer.

5 Laminated structures comprising a plurality of polymeric layers are well known in the art. Such structures are disclosed in U.S. Patent Nos. 4,058,647; 4,198,327; 4,332,858; 4,341,837; and 4,588,648; and European Patent Application No. 0 322 045 A2.

In order for the polymeric laminates to perform successfully, the polymeric layers must be adhered to avoid separation during use. U.S. Patent No. 4,058,647 discloses that ethylene-vinyl alcohol copolymers may be 10 laminated with a polymeric composition comprising both a modified and an unmodified polyolefin and a rubber component. U.S. Patent No. 4,198,327 discloses that polycarbonates and ethylene-vinyl alcohol copolymers may be bonded together with a composition comprising a carboxylated polyolefin and a hydrocarbon elastomer. U.S. Patent Nos. 4,332,858 and 4,341,837 disclose that olefin homopolymers and copolymers may be adhered to polycarbonates using an unhydrogenated block copolymer, such as a maleated block copolymer of styrene and 15 butadiene. U.S. Patent No. 4,588,648 discloses that polypropylene and ethylene-vinyl alcohol copolymers may be laminated with an adhesive layer comprising a grafted copolymer of an olefin and maleic anhydride and an ungrafted polypropylene. European Patent Application No. 0 322 045 A2 discloses that polycarbonate and ethylene vinyl alcohol, for example, may be laminated together using an adhesive composition comprising a carboxylic acid- or acid anhydride-modified polyolefin, such as a maleated polypropylene, and a selectively 20 hydrogenated block copolymer having a block comprising predominantly a hydrogenated conjugated diene such as a saturated styrene-butadiene, or styrene isoprene block copolymer. The use of modified chlorinated, carboxylated polyolefins as primers that produce polymers having improved adhesion to substrates is disclosed in U.S. Patent Nos. 4,954,573, and 4,966,947.

25 None of the above-cited references suggest an appropriate adhesive promoter for a laminate comprising a substantially linear olefin copolymer as described in U.S. Patent Nos. 5,272,236 and 5,278,272. It would be an advance in the art to discover a suitable adhesion promoter for such copolymer.

The present invention is a laminate comprising:

- a) a first substrate comprising a first substantially linear olefin copolymer;
- b) an adhesion promoter superposing the first substrate, which adhesion promoter comprises a polar group 30 functionalized second substantially linear olefin copolymer;
- c) a tackifying resin admixed with the adhesion promoter, or an adhesive superposing the adhesion promoter or admixed therewith; and
- d) a second substrate adhesively bonded to the first substrate;

wherein the first and second substantially linear olefin copolymers are characterized by having:

- i) an  $M_w/M_n$  of less than 3.5;
  - ii) an  $I_{10}/I_2$  of not less than 6; and
  - iii) a critical shear rate at the onset of surface melt fracture of at least 50 percent greater than the critical shear rate at the onset of surface melt fracture of a linear olefin polymer having about the same melt index and
- 5  $M_w/M_n$ ; each substantially linear olefin copolymer being a copolymer of ethylene and a  $C_3$ - $C_{20}$   $\alpha$ -olefin.

In another aspect, the present invention is an adhesion promoter comprising a mixture of:

- a) a copolymer derived from a  $C_2$ - $C_{20}$  olefin and an ethylenically unsaturated carboxylic acid or acid anhydride, and
  - b) a polar group functionalized substantially linear olefin copolymer having:
- 10
  - i) an  $M_w/M_n$  of less than 3.5;
  - ii) an  $I_{10}/I_2$  of not less than 6; and
  - iii) a critical shear rate at the onset of surface melt fracture of at least 50 percent greater than the critical shear rate at the onset of surface melt fracture of a linear olefin polymer having about the same melt index and  $M_w/M_n$ ; each olefin polymer being further characterized as a copolymer of ethylene and a  $C_3$ - $C_{20}$   $\alpha$ -olefin; or

- 15 c) a polar group functionalized copolymer of ethylene and a  $C_3$ - $C_{20}$   $\alpha$ -olefin having a narrow molecular weight distribution, a random distribution of comonomer units along the polymer backbone, and a homogeneity index of at least 75.

- The first substrate of the laminate of the present invention comprises a first substantially linear olefin copolymer. Superposing the first substrate is an adhesion promoter comprising a polar group functionalized
- 20 polymer of a second substantially linear olefin copolymer.

- Alternatively, the adhesion promoter can comprise a polar group functionalized linear copolymer of ethylene and a  $C_3$ - $C_{20}$   $\alpha$ -olefin having a narrow molecular weight distribution, a random distribution of comonomer units along the polymer backbone, and a homogeneity index of at least 75. Such polymers (hereinafter referred to as the '992 polymers) are described by Elston in U.S. Patent No. 3,645,992, and by
- 25 Welborn in U.S. Patent No. 5,324,800, and include those polymers available under the trade name TAFMER<sup>TM</sup> (Trademark of Mitsui Petrochemical) and EXACT<sup>TM</sup> (Trademark of Exxon Chemical).

- The first and second substantially linear olefin copolymers (hereinafter referred to as the substantially linear olefin copolymers) are characterized by having 1) high melt elasticity; 2) high processability; 3) a polydispersity index of less than 3.5; and 4) a melt flow index that is essentially independent of the
- 30 polydispersity index. The substantially linear olefin copolymers are further characterized by having a critical shear rate at the onset of surface melt fracture of at least 50 percent greater than the critical shear rate at the onset of surface melt fracture of a linear olefin polymer having about the same melt index and  $M_w/M_n$ .

The substantially linear olefin copolymers are copolymers of ethylene and at least one C<sub>3</sub>-C<sub>20</sub>  $\alpha$ -olefin, and optionally a C<sub>2</sub>-C<sub>20</sub> acetylenically unsaturated monomer or a C<sub>4</sub>-C<sub>18</sub> diolefin, such as 5-ethylidene-2-norbornene, or an  $\alpha,\omega$ -diene including 1,5-hexadiene, 1,7-octadiene, and 1,9-decadiene. Preferably, the substantially linear olefin copolymers are copolymers of ethylene and a C<sub>4</sub>-C<sub>10</sub>  $\alpha$ -olefin, more preferably ethylene and 1-butene, 1-hexene, 4-methyl-1-pentene, or 1-octene. Most preferably, the substantially linear olefin copolymers are copolymers of ethylene and 1-octene.

The substantially linear olefin copolymers preferably have from 0.01, more preferably from 0.3, to preferably 3, and more preferably to 1, long chain branches per 1000 carbon atoms along the polymer backbone. 10 Long chain branching is defined herein as a chain length of at least about 6 carbon atoms, above which the length cannot be distinguished by carbon NMR spectroscopy. The long chain branch can be about as long as one half the length of the polymer backbone.

The polydispersity index of the substantially linear olefin copolymers (that is, the molecular weight distribution, or the ratio of the weight average molecular weight to the number average molecular weight 15 (M<sub>w</sub>/M<sub>n</sub>)), is less than 3.5, preferably from 1.5 to 2.5. The melt flow index ratio (that is, the I<sub>10</sub>/I<sub>2</sub>, as measured by ASTM D-1238) is at least 5.63, preferably at least 6, more preferably at least 7, and is essentially independent of the polydispersity index, in contrast to conventional polyolefins which show a dependence of the melt flow index on the polydispersity index. This property is illustrated in Figure 2 of U.S. Patent No. 5,272,236, supra. The preparation of substantially linear olefin copolymers is described in detail 20 in the '236 patent.

The density of the substantially linear olefin copolymers is generally in the range of from 0.85 to 0.96, and is preferably in the range of from 0.85, more preferably from 0.86, to 0.89, more preferably to 0.88 g/mL.

The first substrate may optionally contain other conventional additives such as heat stabilizers, weather stabilizers, antistatic agents, nucleating agents, fillers, pigments, dyes, fire retardants, and antiblocking agents. 25 The first substrate may also comprise a mixture of substantially linear olefin polymers, having similar or widely varying weight average (or number average, for that matter) molecular weights. Thus, although substantially linear olefin copolymers have polydispersity indices of less than 3.5, the polydispersity index of the first substrate (as well as the adhesion promoter) is not limited thereto.

The adhesion promoter, which superposes the first substrate, comprises a polar group functionalized 30 second substantially linear olefin copolymer or a polar group functionalized copolymer of ethylene and a '992 polymer. Such a polar group, which is present in sufficient quantities to enhance adhesive properties, is preferably grafted onto a preformed second substantially linear polyolefin or '992 polymer by any suitable means, including means known in the prior art. The polar group may also be incorporated through copolymerization of a suitable monomer containing the desired polar group. Examples of suitable polar groups 35 include halo, particularly chloro and bromo, hydroxyl, carboxyl, carbonyl, phosphono, acid anhydride, amino, epoxy, mercapto, sulfate, sulfonate, amido, and ester groups. Of these, the carboxyl and acid anhydride groups

that are grafted onto the preformed polyolefin are preferred. Examples of unsaturated carboxylic acid and acid anhydride compounds that can be grafted onto the preformed polymer include maleic acid, fumaric acid, itaconic acid, acrylic acid, methacrylic acid, crotonic acid, maleic anhydride, and itaconic anhydride. Maleic anhydride is a preferred grafting compound for the second substantially linear olefin copolymer.

5 Preferably, the polar group is grafted onto a second substantially linear olefin copolymer or '992 polymer which, in its preformed state, is substantially free of polar groups. The term "substantially free" is used herein to refer to a substantially linear olefin copolymer or '992 polymer that contains less than 10 weight percent polar groups based on the weight of the preformed polymer, preferably less than 5 weight percent, more preferably less than 1 weight percent, and most preferably less than 0.1 weight percent. The first substantially linear olefin 10 copolymer is also preferably substantially free of polar groups.

The content of the grafted functional group in the second substantially linear olefin copolymer or a '992 polymer is sufficient to improve the adhesion of the first substrate, and is preferably in the range from 0.05, more preferably from 0.5, and most preferably from 1, to preferably 15, more preferably to 10, and most preferably to 5 weight percent, based on the weight of the second substantially linear olefin copolymer or '992 polymer.

15 The adhesion promoter preferably comprises a mixture of the grafted second substantially linear olefin copolymer or grafted a '992 polymer and a copolymer derived from a) a C<sub>2</sub>-C<sub>20</sub> olefin, preferably a C<sub>2</sub>-C<sub>4</sub> olefin, more preferably ethylene; and b) an unsaturated carboxylic acid or acid anhydride, preferably maleic acid, fumaric acid, itaconic acid, acrylic acid, methacrylic acid, crotonic acid, maleic anhydride, or itaconic anhydride, more preferably acrylic acid or methacrylic acid.

20 Preferably, the grafted second substantially linear olefin copolymer comprises from 10, more preferably from 20, and most preferably from 40 weight percent, to 90, more preferably to 80, and most preferably to 60 weight percent of the adhesion promoter, based on the total weight of the grafted second substantially linear olefin copolymer or grafted '992 polymer and the copolymer derived from the C<sub>2</sub>-C<sub>20</sub> olefin and the unsaturated carboxylic acid or acid anhydride.

25 The laminate is processable (that is, the adhesion promoter promotes adhesion) at a temperature below the melting point of the first substrate. Preferably, the laminate is processable at a temperature from 20°C to 100°C, more preferably to 60°C and most preferably to 40°C.

30 The adhesion promoter may be admixed with a tackifying resin at elevated temperatures, preferably from 100°C to 160°C to form a hot melt adhesive bonding layer between the first and the second substrates. The tackifying resin is compatible with the adhesion promoter and is typically an aliphatic resin, a polyterpene resin, a hydrogenated resin, or a mixed aliphatic aromatic resin. Examples of tackifying resins include those available under the trade name ESCOREZ (Exxon Chemical Co.), PICCOTAC, PICCOVAR, PICCOLYTE (Hercules, Inc.), WINGTAC (Goodyear) and ZONARES (Arizona).

35 Alternatively, the laminate of the present invention may comprise any suitable adhesive that either superposes the adhesion promoter or is admixed therewith. The adhesive may be, for example, neat or solvent-

based, or 1- or 2-part. Examples of suitable adhesives include, but are not restricted to, epoxy resins, urethanes, latexes, acrylates, elastomer-solvent cements, gums, and polysilicones.

A polyisocyanate compound or prepolymer, preferably a di- or triisocyanate compound or prepolymer, such as tris(4-isocyanatophenyl) thiophosphate, toluene diisocyanate, methylene di-p-phenylene isocyanate, or 5 urethane polymers thereof, dissolved in a suitable solvent such as monochlorobenzene or ethyl acetate, can be used to promote solvent-based adhesion. For example, the polyisocyanate compound may be coated over the adhesion promoter layer; formulated with the adhesion promoter, and applied to the first substrate; or formulated with the solvent-based adhesive, and applied to the adhesion promoter layer.

10 The polyisocyanate compound or prepolymer is used in an amount sufficient to promote adhesion between the solvent-based adhesive and the adhesion promoter, preferably from 0.1, more preferably from 0.5, and most preferably from 1, to 25, more preferably to 10, and most preferably to 5 weight percent, based on the weight of the adhesion promoter.

15 Examples of commercially available polyisocyanate compounds or prepolymers include DESMODUR™ RF-E (Trademark of Miles Inc.), PAPI™ polymeric MDI (Trademark of The Dow Chemical Company) and UPACO 3570 (Manufactured by Worthen Industries, UPACO Division).

The isocyanate compound or precursor is also advantageously used in the presence of a catalyst. Suitable catalysts include divalent or tetravalent organotin catalyst such as dimethyltin dilaurate, dimethyltin dicarboxylate, dimethyltin dimercaptide, and stannous octoate; or amine catalysts.

20 The laminate of the present invention further comprises a second substrate adhesively bonded to the first substrate. The second substrate can be any suitable material such as leather, metal, plastic, rubber, glass, wood, or concrete.

The laminate of the present invention is useful for a variety of end-use applications, including bathroom scales, shoe soles, carpet-backing, and automobile windshields.

25 The following examples are for illustrative purposes only and are not intended to limit the scope of the invention.

Example 1 - Preparation and T-Peel Strength of a Polyolefin Elastomer/Leather Laminate

A hot melt adhesive formulation was prepared by mixing together at 150°C until homogeneous, 50 g of ethylene-1-octene copolymer grafted maleic anhydride having a melt flow index of 0.3 g/10 minutes and a density of 0.87 g/mL; and PICCOVAR® AP-10 tackifying resin (50 g, Trademark of Hercules, Inc.).

30 ESCOREZ® 5300 tackifying resin (75 g, Trademark of Exxon Chemical Company) was added to the mixture with stirring until homogeneous. The hot melt adhesive formulation was applied to a strip of ENGAGE® 8200 polyolefin elastomer (Trademark of The Dow Chemical Company). A laminate was prepared for a T-peel test by overlapping the shoe sole compound with a preheated (150°C) abraded leather strip using a 7.6 cm overlap

and a 760  $\mu\text{m}$  bond thickness. The shoe sole strip was 15.2 cm x 2.5 cm x 0.32 cm, and the leather strip was 10 cm x 2.5 cm x 0.095 cm. The adhesive was cured at 100°C for 30 minutes. The excess adhesive around the bonding was removed, and the joints were conditioned at the test temperature for 2 hours prior to testing. The T-peel strength was found to be 34 pli, as measured using an INSTRON® Tensile 4204 Testing System, with a 5 crosshead speed of 25.4 cm/minute, according to ASTM method D1876-72.

**Example 2 - Room-Temperature Cured Epoxy Adhesive for Ethylene-1-Octene Copolymer/E-coated Metal Laminate**

10 Toluene (30.0 g), perchloroethylene (6.0 g), t-butanol (6.0 g), ethylene-1-octene copolymer grafted maleic anhydride having a melt flow index of 0.3 g/10 minutes and a density of 0.87 g/mL (0.34 g), and PRIMACOR® 3460 polymer blend (0.5 g, Trademark of The Dow Chemical Company) were mixed together in a beaker and heated to 80°C until a homogeneous solution was formed. This adhesion promoter blend was cooled to 25°C, then applied as a thin film to a strip of ENGAGE™ 8200 ethylene-1-octene copolymer and allowed to dry at 25°C for 24 hours. A homogeneous mixture of 12.5 g D.E.R.® 331 epoxy resin (Trademark of The Dow Chemical Company), 25.0 g ANCAREZ™ 2364X modifier (Trademark of Air Products and Chemicals, Inc.), 15 and 10.0 g of ANCAMINE™ 2384X curing agent (Trademark of Air Products and Chemicals, Inc.) was applied to the primed strip, and 0.75-mL diameter glass beads were sprinkled on the strip to control thickness. E-coated metal and the ethylene-1-octene copolymer were then mated with a 7.6-cm overlap and the adhesive was cured at 25°C overnight. The T-Peel strength was 40 pli.

20 **Example 3 - Room-Temperature Cured Epoxy Adhesive for Ethylene-1-Octene Copolymer/E-coated Metal Laminate**

The procedure of Example 2 was repeated except that the adhesion promoter blend was a blend of 0.67 g of the ethylene-1-octene copolymer grafted maleic anhydride and 0.17 g of the PRIMACOR® 3460 polymer blend in the toluene/perchloroethylene/t-butanol solvent mixture. The T-Peel strength was 32 pli.

25 **Example 4 - Room-Temperature Cured Epoxy Adhesive for Ethylene-1-Octene Copolymer/E-coated Metal Laminate**

The procedure of Example 2 was repeated except that the adhesion promoter blend was a blend of 0.17 g of the ethylene-1-octene copolymer grafted maleic anhydride and 0.67 g of the PRIMACOR® 3460 polymer blend in the toluene/perchloroethylene/t-butanol solvent mixture. The T-Peel strength was 38 pli.

## WHAT IS CLAIMED IS:

1. A laminate comprising:
  - a) a first substrate comprising a first substantially linear olefin copolymer;
  - b) an adhesion promoter superposing the first substrate, which adhesion promoter comprises a polar group functionalized second substantially linear olefin copolymer;
  - c) a tackifying resin admixed with the adhesion promoter, or an adhesive superposing the adhesion promoter or admixed therewith; and
  - d) a second substrate adhesively bonded to the first substrate;
- wherein the first and second elastic olefin polymers are characterized by having:
  - i) a weight average molecular weight to number average molecular weight ratio of less than 3.5;
  - ii) an  $I_{10}/I_2$  of not less than 6; and
  - iii) a critical shear rate at the onset of surface melt fracture of at least 50 percent greater than the critical shear rate at the onset of surface melt fracture of a linear olefin polymer having about the same melt index and  $M_w/M_n$ ; each olefin polymer being further characterized as a copolymer of ethylene and a  $C_3$ - $C_{20}$   $\alpha$ -olefin.
- 15 2. The laminate of Claim 1 wherein the adhesion promoter further comprises a copolymer derived from a  $C_2$ - $C_{20}$  olefin and an ethylenically unsaturated carboxylic acid or acid anhydride.
3. The laminate of Claim 2 wherein the copolymer of the olefin and the ethylenically unsaturated carboxylic acid is a copolymer of ethylene and acrylic acid, or ethylene and methacrylic acid.
4. The laminate of any of Claims 1 to 3 wherein each substantially linear olefin copolymer is independently a copolymer of ethylene and 1-butene, 4-methyl-1-pentene, 1-hexene, or 1-octene.
- 20 5. The laminate of any of Claims 1 to 4 wherein each substantially linear olefin copolymer is a copolymer of ethylene and 1-octene having a density of from 0.86 to 0.88 g/mL.
6. The laminate of any of Claims 1 to 5 wherein the polar group functionalized second substantially linear olefin copolymer comprises an acid anhydride group or a carboxylic acid group grafted onto an ethylene-1-octene copolymer which, in the preformed state, is substantially free of polar groups, wherein the polar group functionalized polymer comprises 0.05 to 10 weight percent of maleic anhydride grafted onto the preformed ethylene-1-octene copolymer, based on the weight of the preformed ethylene-1-octene copolymer and wherein the ratio of the maleic anhydride grafted ethylene-1-octene copolymer to the copolymer of ethylene and acrylic acid, or ethylene and methacrylic acid is in the range of 20:80 to 80:20.
- 25 30 7. The laminate of any of Claims 1 to 6 wherein a solvent-based adhesive formulation superposes the adhesion promoter or is admixed therewith and wherein the adhesion promoter or the solvent-based adhesive further comprises an isocyanate compound or isocyanate prepolymer.

8. The laminate of any of Claims 1 to 9 wherein the second substrate comprises a metal, glass, wood, cement, plastic, rubber, or leather.

9. An adhesion promoter comprising a mixture of:

- a) a copolymer derived from a C<sub>2</sub>-C<sub>20</sub> olefin and an ethylenically unsaturated carboxylic acid or acid anhydride, and
- 5 b) a polar group functionalized substantially linear olefin copolymer having:
  - i) an M<sub>w</sub>/M<sub>n</sub> of less than 3.5;
  - ii) an I<sub>10</sub>/I<sub>2</sub> of not less than 6; and
  - iii) a critical shear rate at the onset of surface melt fracture of at least 50 percent greater than the critical
- 10 shear rate at the onset of surface melt fracture of a linear olefin polymer having about the same melt index and M<sub>w</sub>/M<sub>n</sub>; each olefin polymer being further characterized as a copolymer of ethylene and a C<sub>3</sub>-C<sub>20</sub>  $\alpha$ -olefin; or
- c) a polar group functionalized linear olefin copolymer having a narrow molecular weight distribution; a random distribution of comonomer units along the polymer backbone and a homogeneity index of at least 75.

10. An adhesion promoter which comprises a mixture of:

- 15 a) a copolymer of a C<sub>2</sub>-C<sub>20</sub> olefin and an ethylenically unsaturated carboxylic acid or acid anhydride, and
- b) a polar group functionalized substantially linear olefin copolymer having:
  - i) an M<sub>w</sub>/M<sub>n</sub> of less than 3.5;
  - ii) an I<sub>10</sub>/I<sub>2</sub> of not less than 6; and
  - iii) a critical shear rate at the onset of surface melt fracture of at least 50 percent greater than the critical
- 20 shear rate at the onset of surface melt fracture of a linear olefin polymer having about the same melt index and M<sub>w</sub>/M<sub>n</sub>; each olefin polymer being further characterized as a copolymer of ethylene and a C<sub>3</sub>-C<sub>20</sub>  $\alpha$ -olefin, wherein the copolymer of the C<sub>2</sub>-C<sub>20</sub> olefin and the ethylenically unsaturated carboxylic acid comprises a copolymer of ethylene and acrylic acid or methacrylic acid, and the polar group functionalized substantially linear olefin copolymer comprises maleic anhydride grafted onto an ethylene-1-octene copolymer which, in the preformed state, is substantially free of polar groups and has a density of from 0.86 to 0.88 g/mL, and wherein the ratio of the maleic anhydride grafted ethylene-1-octene copolymer to the copolymer of ethylene and acrylic acid or ethylene and methacrylic acid is in the range of 20:80 to 80:20.
- 25

# INTERNATIONAL SEARCH REPORT

Intern. Application No  
PCT/US 96/12289

**A. CLASSIFICATION OF SUBJECT MATTER**  
IPC 6 B32B27/32 C09J151/06

According to International Patent Classification (IPC) or to both national classification and IPC

**B. FIELDS SEARCHED**

Minimum documentation searched (classification system followed by classification symbols)  
IPC 6 B32B C09J

Documentation searched other than minimum documentation to the extent that such documents are included in the fields searched

Electronic data base consulted during the international search (name of data base and, where practical, search terms used)

**C. DOCUMENTS CONSIDERED TO BE RELEVANT**

Category	Citation of document, with indication, where appropriate, of the relevant passages	Relevant to claim No.
A	DATABASE WPI Section Ch, Week 9434 Derwent Publications Ltd., London, GB; Class A17, AN 94-275906 XP002018841 & JP,A,06 206 947 (MITSUI PETROCHEM IND CO LTD) , 26 July 1994 see abstract ---	1,9,10
A	GB,A,2 116 187 (ACC CHEM CO;GETTY CHEMICAL CO) 21 September 1983 see page 1, line 1 - line 32; claims ---	1,9,10
A	GB,A,2 113 696 (ACC CHEM CO;GETTY CHEMICAL CO) 10 August 1983 see page 1, line 5 - line 6 see page 1, line 17 - line 44 see page 2, line 22 - line 33 ---	1,9,10
	-/-	

Further documents are listed in the continuation of box C.

Patent family members are listed in annex.

\* Special categories of cited documents :

- \*A\* document defining the general state of the art which is not considered to be of particular relevance
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Date of the actual completion of the international search	Date of mailing of the international search report
19 November 1996	29.11.96
Name and mailing address of the ISA European Patent Office, P.B. 5818 Patentlaan 2 NL - 2280 HV Rijswijk Tel. (+ 31-70) 340-2040, Tx. 31 651 cpo nl, Fax (+ 31-70) 340-3016	Authorized officer  De Jonge, S

# INTERNATIONAL SEARCH REPORT

Intern: Application No  
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C(Continuation) DOCUMENTS CONSIDERED TO BE RELEVANT		
Category *	Citation of document, with indication, where appropriate, of the relevant passages	Relevant to claim No.
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